Coherent backscattering by two-sphere clusters

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Rigorous numerical solutions of Maxwell's equations are used to show, for what is believed to be the first time, that simple scattering systems composed of two interacting wavelength-sized spheres exhibit a coherent backscattering effect analogous to that observed previously for optically thick discrete random media comprising large numbers of scatterers. © 1996 Optical Society of America

The phenomenon of enhanced backscattering of light by discrete random media has been extensively studied during the past decade both experimentally and theoretically[1] and has been found important in explaining opposition scattering effects observed for solar system bodies.[2] This phenomenon is caused by constructive interference of two waves traveling along the same scattering path but in opposite directions and arriving at the backscattering direction with the same phase. It was realized recently that enhanced backscattering can be produced not only by ensembles comprising large numbers of scatterers but also by clusters composed of two scattering particles. However, because of theoretical difficulties in solving Maxwell's equations for closely packed clusters of wavelength-sized particles, the problem has been studied for only the simplest case of two dipoles, in which case the amplitude of the coherent backscattering intensity peak was extremely small and, thus, essentially unobservable.[1,3,4]

In a recent Letter Mishchenko and Mackowski developed an efficient T-matrix method for rigorously computing light scattering by randomly oriented two-sphere clusters with component-sphere sizes comparable with the wavelength of the incident light.[5] Therefore it is my aim in this Letter to describe an application of this computational technique to a search of coherent backscattering for two-sphere clusters consisting of wavelength-sized components.

Qualitative physical considerations predict two primary interference peaks in the intensity scattered by a two-sphere cluster. The forward-scattering peak is produced by interference of the waves singly scattered by the component spheres.[6] Because for the exactly forward-scattering direction the interference is constructive regardless of the distance d between the centers of the component spheres and the cluster orientation, the forward-scattered intensity should be roughly doubled compared with that for two noninteracting spheres.[1,7,8] The backscattering peak is produced primarily by interference of two waves scattered along the following paths: source of light $\rightarrow$ sphere 1 $\rightarrow$ sphere 2 $\rightarrow$ detector and source of light $\rightarrow$ sphere 2 $\rightarrow$ sphere 1 $\rightarrow$ detector.[1] For the exact backscattering direction the interference of the two waves is always constructive and should cause a distinct backscattering enhancement of intensity. However, because this is a second-order-scattering phenomenon, one should expect a much weaker effect than in the forward direction. The angular widths of both peaks should be of the order of $1/kd$, where $k = 2\pi/\lambda$ is the wave number and λ is the wavelength of the incident light.

In these simplistic physical considerations several important factors that make the detection of the interference peaks difficult are not taken into account (cf. Ref. 9). Indeed, the scattering pattern for a monodisperse two-sphere cluster in a fixed orientation and with a fixed d is heavily burdened by secondary interference maxima[10,11] and by the complicated resonance structure typical of single monodisperse spheres.[11,12] The interference pattern is further complicated by the near-field effects that result from component spheres in a closely packed cluster not being in the far-field zones of each other.[11] To smooth the effects of these factors out and make the weak backscattering peak detectable, the scattering pattern should be averaged over sphere sizes, cluster orientations, and distances between the component spheres. Furthermore, for peaks that are narrow and well distinguishable the average distance between the centers of the component spheres must be much larger than the wavelength: $\langle kd \rangle \gg 1$.[4,8]

The method described in Ref. 5 makes possible efficient and numerically accurate computation of the differential scattering cross section σ for a randomly oriented two-sphere cluster with an arbitrary distance between the component spheres. σ describes the angular distribution of the scattered intensity in the far-field zone of the cluster, provided that the incident beam is unpolarized.[7] We assume that the spheres that form a two-sphere cluster have the same size and average σ over sizes of component spheres and distances d between their centers. In ensemble averaging, we use the standard gamma distribution of component-sphere size parameters x,

$$n(x) \propto x^{(1-3\nu_{\mathrm{eff}})/\nu_{\mathrm{eff}}} \exp[-x/(r_{\mathrm{eff}}\nu_{\mathrm{eff}})], \tag{1}$$

where $x = kr$, r is component-sphere radius, $n(x)\mathrm{d}x$ is the fraction of spheres with size parameters from x to $x + \mathrm{d}x$, x_{eff} is the cross-section-area-weighted effective size parameter, and the effective variance ν_{eff} provides a measure of the width of the distribution.[12] It is assumed that d is equal to n times the component-sphere radius so that the effective distance parameter $(kd)_{\mathrm{eff}}$ is equal to nx_{eff}.

In the absence of electromagnetic interactions between component spheres (i.e., with interference and near-field multiple-scattering effects turned off), the differential scattering cross section for a two-sphere cluster would be exactly equal to twice that for a single sphere: $\sigma_{\text{cluster}} = 2\sigma_{\text{sphere}}$. Therefore it is convenient for one to demonstrate the effect of electromagnetic interactions by plotting the normalized differential scattering cross section $\sigma^* = \sigma_{\text{cluster}}/(2\sigma_{\text{sphere}})$ versus scattering angle Θ and looking for deviations of σ^* from 1. Figure 1 shows σ^* for randomly oriented polydisperse clusters with $x_{\text{eff}} = 5$, $\nu_{\text{eff}} = 0.05$, $(kd)_{\text{eff}} = 25$, and refractive indices $m_r = 1.2$ and $m_r = 1.5$. Despite some minor differences, the two curves are remarkably similar, especially at forward-scattering and backscattering angles. Both curves show the expected strong forward-scattering peak with an angular width of the order of $1/(kd)_{\text{eff}}$ and an amplitude close to 2. The deviation of $\sigma^*(0°)$ from 2 is apparently caused by near-field effects. The same effects cause the $\sigma^*(180°)$ value for the refractive index 1.5 to be less than 1. However, both curves exhibit distinct backscattering peaks superposed on a relatively smooth background. We have repeated these calculations for x_{eff} varying from 3 to 6, $(kd)_{\text{eff}}$ varying from 20 to 30, and m_r varying from 1.2 to 1.5. In all cases the σ^* curves were similar to those depicted in Fig. 1 and showed a strong forward-scattering peak and a small-amplitude but distinct backscattering peak. The persistence of this general pattern with varying x_{eff}, $(kd)_{\text{eff}}$, and m_r strongly indicates that it is caused by interference.

That the forward-scattering peak in the σ^* curve is produced by interference can be verified with a plot of σ^* versus the dimensionless angular parameter $u = 2(kd)_{\text{eff}} \sin(\Theta/2)$.[8] Indeed, if interference is the primary effect, the forward-scattering profile of σ^* as a function of u should be essentially the same for equally sized spheres regardless of $(kd)_{\text{eff}}$. Figure 2 shows calculations for $x_{\text{eff}} = 5$, $\nu_{\text{eff}} = 0.05$, $m_r = 1.2$, and $(kd)_{\text{eff}} = 20, 25, 30$. As expected, all three curves almost coincide. The amplitude of the peak decreases slightly with decreasing $(kd)_{\text{eff}}$, thus indicating the increasing negative influence of the near-field effects. These effects can be interpreted in part as mutual shadowing of component spheres when light is incident upon or scattered nearly parallel to the cluster axis, or both.

Figure 3 shows the normalized backscattered intensity $\sigma^*(\Theta)/\sigma^*(180°)$ versus the parameter $w = 2(kd)_{\text{eff}} \sin[(\pi - \Theta)/2]$ for the same cluster parameters. As is the case for the forward-scattering peak, the angular width of the backscattering peak in units of w is nearly independent of $(kd)_{\text{eff}}$, thus pointing to coherent backscattering as its cause. It is important to emphasize, however, that despite their common interference nature the angular widths of the forward-scattering and the backscattering peaks are not necessarily equal. Indeed, for a two-sphere cluster in a fixed orientation with respect to the incident beam the angular widths of both peaks strongly depend on the cluster orientation and are minimal when the cluster axis is perpendicular to the direction of incidence and maximal when the cluster axis is parallel to this direction (nose-on orientation). Because of strongly anisotropic scattering by individual wavelength-sized spheres, the relative contributions of different orientations to the forward-scattered and the backscattered peaks for a randomly oriented cluster can be substantially different, thus causing different angular widths of the peaks, as is demonstrated in Figs. 2 and 3. Furthermore, with decreasing distance between the component spheres the increasing effect of mutual shadowing can weaken the relative contributions of near nose-on orientations and thus reduce the width of the backscattering peak. This can indeed be seen in Fig. 3. It should also be noted that the degree of scattering anisotropy for individual spheres is refractive index dependent. This can, apparently, explain the somewhat different widths of the forward-scattering as well as the backscattering peaks depicted in Fig. 1

Figure 3 shows that the vertex of the backscattering peak is rounded off, which is explained by confined geometry of light scattering by two-sphere clusters and the absence of infinitely long photon paths.[1] The amplitude of the backscattering peak increases with decreasing distance between the component spheres, which is also consistent with the explanation in terms of coherent backscattering. Indeed,

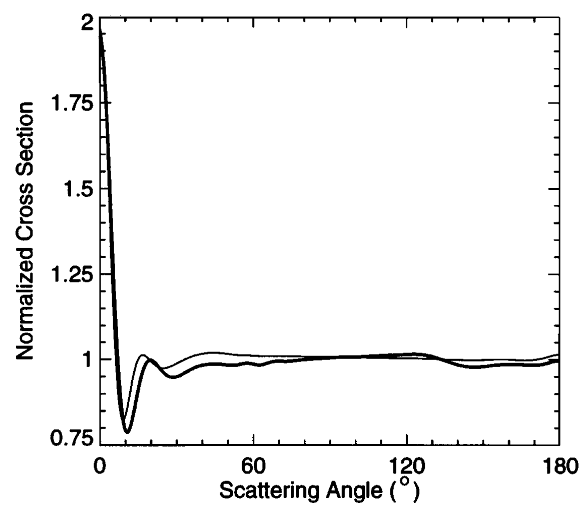

Fig. 1. σ^* versus Θ for polydisperse, randomly oriented two-sphere clusters with $x_{\text{eff}} = 5$, $\nu_{\text{eff}} = 0.05$, $(kd)_{\text{eff}} = 25$, $m_r = 1.2$ (thin solid curve), $m_r = 1.5$ (thick solid curve).

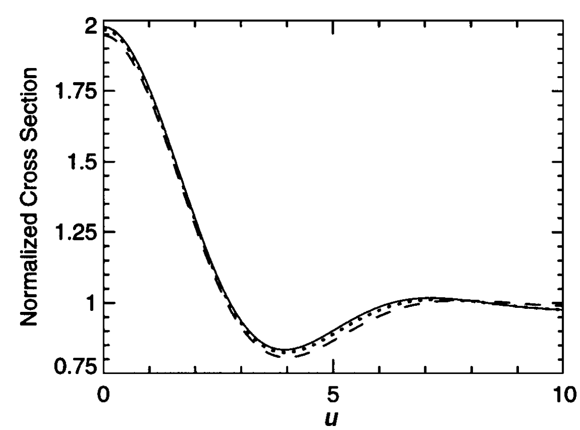

Fig. 2. σ^* versus u for polydisperse, randomly oriented two-sphere clusters with $x_{\text{eff}} = 5$, $\nu_{\text{eff}} = 0.05$, $m_r = 1.2$, and $(kd)_{\text{eff}} = 30$ (solid curve), 25 (dotted curve), and 20 (dashed curve).

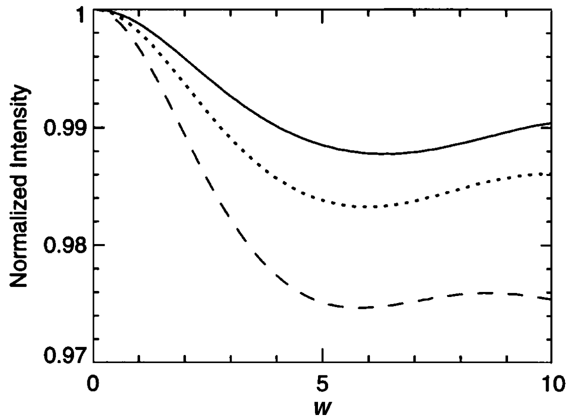

Fig. 3. As in Fig. 2, but for normalized backscattered intensity versus w.

unlike in the case with the forward-scattering enhancement, which results from constructive interference of singly scattered waves and is essentially independent of the distance between the component spheres, the backscattering intensity peak is caused by interference of double-scattered waves and weakens proportionally to d^{-2}. For all cases shown in Fig. 3, the amplitude of the backscattering peak is much smaller than typical values computed for media composed of large numbers of scatterers.[13] This is an expected result because with an increasing number of particles N the contribution of single-scattered light is proportional to N, whereas the contribution of double-scattered light rises as $N(N-1)$.

Another manifestation of coherent backscattering for discrete random media illuminated by unpolarized light is the so-called polarization opposition effect observed as a narrow branch of negative polarization at scattering angles close to 180°.[14] This phenomenon is equivalent to what is called spatial anisotropy of the polarized cone of enhanced backscattering in the case of illumination of the medium by a linearly polarized beam.[15] However, this effect is rather weak even for an optically semi-infinite medium composed of strongly polarizing Rayleigh scatterers[1,14,15] and is expected to be much weaker for two-sphere clusters composed of wavelength-sized particles. Therefore, even if it was present in our computations for two-sphere clusters, it was masked by a much stronger single-sphere polarization and therefore was essentially indistinguishable.

In summary, the exact and efficient T-matrix method developed in Ref. 5 has been used for extensive computation of light scattering by polydisperse, randomly oriented two-sphere clusters with well-separated, wavelength-sized components. Our computations have shown that these simple scattering systems exhibit not only the well-known forward-scattering intensity peak caused by constructive interference of single-scattered waves[7,8] but also distinct enhancement of backscattered intensity resulting from interference of waves scattered twice. The latter phenomenon is equivalent to that observed earlier for optically thick discrete random media and called coherent backscattering.

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References

1. Yu. N. Barabanenkov, Yu. A. Kravtsov, V. D. Ozrin, and A. I. Saichev, in *Progress in Optics XXIX*, E. Wolf, ed. (Elsevier, New York, 1991), p. 65; B. A. van Tiggelen, "Multiple scattering and localization of light," Ph.D. dissertation (Amsterdam University, Amsterdam, The Netherlands, 1992).
2. B. Hapke, Icarus **88,** 407 (1990); S. J. Ostro, Rev. Mod. Phys. **65,** 1235 (1993); M. I. Mishchenko and J. M. Dlugach, Planet. Space Sci. **41,** 173 (1993); K. Muinonen, in *Asteroids, Comets, Meteors 1993, Proceedings of the 160th Symposium of the International Astronomical Union* (Kluwer, Dordrecht, The Netherlands, 1994), p. 271.
3. K. Muinonen, "Light scattering by inhomogeneous media: backward enhancement and reversal of linear polarization," Ph.D. dissertation (University of Helsinki, Helsinki, Finland, 1990).
4. F. M. Ismagilov and Yu. A. Kravtsov, Waves Random Med. **3,** 17 (1993).
5. M. I. Mishchenko and D. W. Mackowski, Opt. Lett. **19,** 1604 (1994). Note that the second sentence of the last paragraph on page 1605 should read as follows: "The index of refraction of the spheres is $1.5 + 0.02i$ and the size parameter of each sphere is 15.874."
6. The interference forward-scattering peak should not be confused with the forward-scattering peak produced by the diffraction of light on the particle cross section if particle size is comparable with or larger than a wavelength.
7. H. C. van de Hulst, *Light Scattering by Small Particles* (Wiley, New York, 1957); C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1983).
8. F. M. Ismagilov and Yu. A. Kravtsov, Phys. Lett. A **188,** 91 (1994).
9. V. P. Tishkovets, Kinem. Fiz. Nebes. Tel **10**(2), 58–63 (1994).
10. K. A. Fuller, G. W. Kattawar, and R. T. Wang, Appl. Opt. **25,** 2521 (1986).
11. M. I. Mishchenko, D. W. Mackowski, and L. D. Travis, Appl. Opt. **34,** 4589 (1995).
12. J. E. Hansen and L. D. Travis, Space Sci. Rev. **16,** 527 (1974).
13. M. I. Mishchenko, J. Opt. Soc. Am. A **9,** 978 (1992).
14. M. I. Mishchenko, Astrophys. J. **411,** 351 (1993).
15. M. P. van Albada, M. B. van der Mark, and A. Lagendijk, J. Phys. D **21,** S28 (1988).